

***AEROSOL COMPOSITION, CHEMISTRY, AND SOURCE CHARACTERIZATION
DURING THE 2008 VOCALS EXPERIMENT***

Y.-N. Lee¹, S. Springston¹, J. Jayne², J. Wang¹, G. Senum¹, J. Hubbe³, M. Alexander³,
J. Brioude⁴, S. Spak⁵, M. Mena⁶, L. Kleinman¹, P. Daum¹

¹Brookhaven National Laboratory; ²Aerodyne Research Inc., ³Pacific Northwest National Laboratory;
⁴National Oceanographic and Atmospheric Administration; ⁵University of Iowa; ⁶Universidad Andrés Bello

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Environmental Sciences Department/Atmospheric Sciences Division
Brookhaven National Laboratory
P.O. Box, Upton, NY
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ABSTRACT

Chemical composition of fine aerosol particles over the northern Chilean coastal waters was determined on board the US DOE G-1 aircraft during the VOCALS (VAMOS Ocean-Cloud-Atmosphere-Land Study) field experiment between October 16 and November 15, 2008. Chemical species determined included SO_4^{2-} , NO_3^- , NH_4^+ , and total organics (Org) using an Aerodyne Aerosol Mass Spectrometer, and SO_4^{2-} , NO_3^- , NH_4^+ , Na^+ , Cl^- , CH_3SO_3^- , Mg^{2+} , Ca^{2+} , and K^+ using a particle-into-liquid sampler-ion chromatography technique. The results show the marine boundary layer (MBL) aerosol mass was dominated by non-sea-salt SO_4^{2-} followed by Na^+ , Cl^- , Org, NO_3^- , and NH_4^+ , in decreasing importance; CH_3SO_3^- , Ca^{2+} , and K^+ rarely exceeded their respective limits of detection. The SO_4^{2-} aerosols were strongly acidic as the equivalent NH_4^+ to SO_4^{2-} ratio was only ~ 0.25 on average. NaCl particles, presumably of sea-salt origin, showed chloride deficits but retained Cl^- typically more than half the equivalency of Na^+ , and are believed to be externally mixed with the acidic sulfate aerosols. Nitrate was observed only on sea-salt particles, consistent with adsorption of HNO_3 on non-acidic sea-salt aerosols, responsible partly for the Cl^- deficit. Dust particles appeared to play a minor role judging from the small volume differences between that derived from the observed mass concentrations and that calculated based on particle size distributions. Because SO_4^{2-} concentrations in the study domain were substantial (~ 0.5 - $\sim 3 \mu\text{g}/\text{m}^3$) with a strong gradient (highest near the shore decreasing with distance from land), and the ocean-emitted dimethylsulfide and its unique oxidation product, CH_3SO_3^- , were very low (i.e., ≤ 40 parts per trillion and $< 0.05 \mu\text{g}/\text{m}^3$, respectively), the observed SO_4^{2-} aerosols are believed to be primarily of terrestrial origin. Back trajectory calculations indicate sulfur emissions from smelters and power plants along coastal regions of Peru and Chile are the main sources of these SO_4^{2-} aerosols. However, compared to observations, model calculations appeared to underestimate sulfate concentrations based on an existing emission inventory. An up-to-date and comprehensive emission inventory currently being constructed will be tested to examine models' capability in predicting distributions of sulfate precursors and other emitted materials such as CO both in the MBL and the free troposphere by comparing to observations. Analysis of the role sea-salt particles play in in-cloud SO_4^{2-} production and in acidification of seawater by deposition will also be presented.